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## Polar vortex evolution during the 2002 Antarctic major warming as observed by the Odin satellite

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[1] In September 2002 the Antarctic polar vortex split in two under the influence of a sudden warming. During this event, the Odin satellite was able to measure both ozone ( $O_3$ ) and chlorine monoxide (ClO), a key constituent responsible for the so-called “ozone hole”, together with nitrous oxide ( $N_2O$ ), a dynamical tracer, and nitric acid ( $HNO_3$ ) and nitrogen dioxide ( $NO_2$ ), tracers of denitrification. The submillimeter radiometer (SMR) microwave instrument and the Optical Spectrograph and Infrared Imager System (OSIRIS) UV-visible light spectrometer (VIS) and IR instrument on board Odin have sounded the polar vortex during three different periods: before (19–20 September), during (24–25 September), and after (1–2 and 4–5 October) the vortex split. Odin observations coupled with the Reactive Processes Ruling the Ozone Budget in the Stratosphere (REPROBUS) chemical transport model at and above 500 K isentropic surfaces (heights above 18 km) reveal that on 19–20 September the Antarctic vortex was dynamically stable and chemically nominal: denitrified, with a nearly complete chlorine activation, and a 70%  $O_3$  loss at 500 K. On 25–26 September the unusual morphology of the vortex is monitored by the  $N_2O$  observations. The measured ClO decay is consistent with other observations performed in 2002 and in the past. The vortex split episode is followed by a nearly complete deactivation of the ClO radicals on 1–2 October, leading to the end of the chemical  $O_3$  loss, while  $HNO_3$  and  $NO_2$  fields start increasing. This acceleration of the chlorine deactivation results from the warming of the Antarctic vortex in 2002, putting an early end to the polar stratospheric cloud season. The model simulation suggests that the vortex elongation toward regions of strong solar irradiance also favored the rapid reformation of ClONO<sub>2</sub>. The observed dynamical and chemical evolution of the 2002 polar vortex is qualitatively well reproduced by REPROBUS. Quantitative differences

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are mainly attributable to the too weak amounts of  $\text{HNO}_3$  in the model, which do not produce enough  $\text{NO}_2$  in presence of sunlight to deactivate chlorine as fast as observed by Odin.

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## 1. Introduction

[2] The Antarctic early spring is usually characterized by a cold and stable stratospheric vortex, with elevated amounts of active chlorine and ongoing ozone destruction [World Meteorological Organization, 1998]. In September 2002 an unusual event occurred in the stratosphere over Antarctica. At isentropic levels above about 600 K, the polar vortex split in two under the influence of a sudden stratospheric warming. This phenomenon had never been observed before in the Southern Hemisphere [see, e.g., Simmons *et al.*, 2003] and has been recently studied in several articles dealing with the dynamical evolution of the vortex [e.g., Manney *et al.*, 2005] or the evolution of the ozone field [e.g., Hoppel *et al.*, 2003].

[3] During this period, the Odin satellite was able to monitor both the evolutions of ozone ( $\text{O}_3$ ) and chlorine monoxide (ClO), the key constituent for destroying ozone in polar vortices, producing the so-called “ozone hole” [Farman *et al.*, 1985]. In addition, Odin has observed other chemical species related to polar ozone depletion, such as nitric acid ( $\text{HNO}_3$ ) and nitrogen dioxide ( $\text{NO}_2$ ), as well as the dynamical evolution of the vortex that can be inferred from the measurements of nitrous oxide ( $\text{N}_2\text{O}$ ). These measurements combined with the calculations from the three-dimensional (3-D) chemical transport model REPROBUS allow the characterization of the chemical and dynamical evolution of the Southern Hemisphere vortex in late September to early October 2002. We particularly focused on four periods: 19–20 September 2002, prior to the vortex split; 25–26 September 2002, during the vortex split; and 1–2 and 4–5 October 2002, after the vortex split.

[4] Section 2 deals with the Odin measurements in terms of sensitivity, horizontal and vertical resolutions, vertical coverage and errors. Section 3 briefly presents the calculations performed with the three-dimensional (3-D) model. The detailed description of the vortex evolution in terms of chemistry and dynamics, as observed by Odin and as calculated by REPROBUS, is presented in section 4.

## 2. Odin Measurements

[5] The Odin minisatellite is a Swedish-led project funded jointly by Sweden, Canada, France, and Finland [Murtagh *et al.*, 2002; Nordh *et al.*, 2003]. It was placed into a 600-km Sun-synchronous, terminator orbit by a START-1 rocket on 20 February 2001 from Svobodny, Russia. Odin includes two instruments. A UV-visible and infrared instrument, the Optical Spectrograph and Infrared Imager System (OSIRIS), that is capable of detecting  $\text{O}_3$ ,  $\text{NO}_2$ , and aerosol in the scattered sunlight in the wavelength range 280–800 nm and the related oxygen airglow emission at 1.27 mm [Llewellyn *et al.*, 2004]. The

second instrument is the submillimeter radiometer (SMR) [Frisk *et al.*, 2003] that can simultaneously measure  $\text{O}_3$ , ClO and  $\text{N}_2\text{O}$  in the frequency domain 501.18–502.38 GHz, together with  $\text{HNO}_3$  at 544.20–545.00 GHz. Other molecules such as water vapor ( $\text{H}_2\text{O}$ ) and its isotopes, as well as carbon monoxide (CO) [Dupuy *et al.*, 2004], can also be detected in the stratosphere and in the mesosphere. It should be noted that the microwave measurements are not degraded by the presence of Polar Stratospheric Clouds (PSCs) and aerosols in the stratosphere.

[6] In the basic stratospheric observation mode, the Earth's limb is scanned from 7 to 70 km in about 90 s. The horizontal resolution of Odin measurements along the orbit plane is  $\sim 500$  km. The SMR data analysis uses the optimal estimation method [Rodgers, 1976] to retrieve vertical profiles of the atmospheric constituents having lines in the frequency band under consideration [Baron *et al.*, 2002]. For OSIRIS, global number density profiles of  $\text{O}_3$  and  $\text{NO}_2$  are retrieved from limb-scattered sunlight measurements using the maximum a posteriori estimator method [Rodgers, 2000], with some intermediate spectral analysis. For  $\text{O}_3$ , measurements at three wavelengths in the Chappuis region are combined [von Savigny *et al.*, 2003] and for  $\text{NO}_2$ , differential optical absorption spectroscopy (DOAS) is applied in the wavelength window 435–451 nm [Haley *et al.*, 2004]. Data processing for both instruments uses temperature and pressure fields analyzed by the European Centre for Medium-range Weather Forecast (ECMWF). The vertical resolution of the measurements depends on the species and the integration time versus altitude domain considered. For SMR [Urban *et al.*, 2004], at polar latitudes, measured ClO profiles cover the altitude range 17–50 km, with a vertical resolution of typically 2–2.5 km and a corresponding single-scan retrieval precision of about 0.15–0.20 ppbv.  $\text{N}_2\text{O}$  is retrieved above 14 km with a vertical resolution of 2 km and a precision in the order of 5% (10–20 ppbv). The SMR measurement range for  $\text{O}_3$  is 20–50 km in the 501 GHz band, with a resolution of around 2.5 km and a single-scan precision in the order of 25% (0.5–1.5 ppmv). The  $\text{HNO}_3$  profile can be retrieved from 20 to 35 km with a resolution of 2 km, and a single-scan precision of 1–1.5 ppbv. Upper limits for the systematic errors of the measurements in the lower stratosphere can be estimated to be about 0.2 ppbv for ClO, 25 ppbv for  $\text{N}_2\text{O}$ , and 1 ppmv for  $\text{O}_3$ . Systematic errors for  $\text{HNO}_3$  are not clearly determined yet from our preliminary validation studies. For OSIRIS, the accuracy of the ozone retrievals is estimated to be better than 10% over an altitude range of 15 to 35 km with a resolution of about 2 km [Petelina *et al.*, 2004; von Savigny *et al.*, 2005]. The  $\text{NO}_2$  vertical profiles can be retrieved over an

altitude range of 15 to 40 km with a resolution of 2–3 km and an accuracy of about 10% at the peak [Haley *et al.*, 2004].

### 3. Model Calculations

[7] Three-dimensional simulations of the 2002 ozone hole have been performed with the REPROBUS chemical transport model [Lefèvre *et al.*, 1994, 1998] which has been widely used in previous studies of the stratospheric chemistry [e.g., Lefèvre *et al.*, 1994; Deniel *et al.*, 1998; Hoppel *et al.*, 2002]. The model is designed to perform annual simulations as well as detailed process studies. It computes the evolution of 55 species by means of about 160 gas-phase and heterogeneous reactions, with a time step of 15 min in this study. A semi-Lagrangian code transports 40 species or chemical families, typically long-lived tracers but also more unstable compounds which may have a long lifetime in darkness. Kinetics parameters used in the present study are based on the most recent data [Sander *et al.*, 2003]. The new laboratory measurements of photodissociation cross sections of HO<sub>2</sub>NO<sub>2</sub> both in the UV [Knight *et al.*, 2002] and in the near IR [Roehl *et al.*, 2002] have been included in the photodissociation calculations. The heterogeneous chemistry module used in the present simulation includes reactions on binary and ternary liquid aerosols, as well as on water-ice particles. The composition and volume of the liquid droplets are computed from the temperature and pressure conditions and the available amounts of H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>, and H<sub>2</sub>O [Carslaw *et al.*, 1995]. Sedimentation of polar stratospheric cloud particles is triggered in the model when the temperature reaches the ice frost point. The ice particles are assumed to include HNO<sub>3</sub> in the form of nitric acid trihydrate (NAT). Their mean radius is calculated as a function of the amount of condensed water and a density of  $5 \times 10^{-3}$  particles/cm<sup>3</sup>.

[8] REPROBUS was integrated from 1 April 2002 to 15 October 2002. Temperatures, winds and ground pressure were specified from the 6-hourly ECMWF operational analysis. The model extends from the surface up to 0.1 hPa on 42 levels, resulting in a vertical resolution of about 1.3 km in the lower stratosphere. The horizontal resolution is 2° latitude by 2° longitude. The ozone field was initialized on 1 April 2002 from the three-dimensional ECMWF ozone analysis. Other species were initialized from an April zonal mean computed from a long-term simulation of REPROBUS. Model profiles were extracted online during the simulation to coincide to the time and location of the Odin measurements.

### 4. Vortex Evolution

[9] We focus here on four periods: 19–20 September 2002, 25–26 September 2002, 1–2 October 2002, and 4–5 October 2002. Each period corresponds to measurements performed over 24 hours starting from 1200 UT and includes about 14 orbits. Figures 1, 3, 5, and 7 show O<sub>3</sub>, ClO, N<sub>2</sub>O, HNO<sub>3</sub>, as well as sunrise and sunset NO<sub>2</sub> linearly interpolated onto the 500 K potential temperature surface, corresponding to an altitude range from ~18 to ~22 km for latitudes poleward of 30°S for the corresponding four periods. Data have been triangularly interpolated into bins of

5° wide in latitude and 10° wide in longitude. The fields for ClO, N<sub>2</sub>O, and HNO<sub>3</sub> are from SMR measurements. The fields for O<sub>3</sub> and NO<sub>2</sub> are from OSIRIS, except on 19–20 September when solar zenith angles (SZA) larger than 90° in the Antarctic region precluded optical measurements. The SMR ozone is displayed for that date. For each period, Figures 2, 4, 6, and 8 show a latitude versus height representation of the Odin measurements along one selected orbit (indicated by a thick black line on Figures 1, 3, 5, and 7). Odin measurements are compared to the REPROBUS results coincident in time and location. Species not measured by Odin (HCl and ClONO<sub>2</sub>) and additional diagnostics (chlorine activation ratio, ozone loss, SZA) together with ECMWF temperature fields are also presented at 500 K.

#### 4.1. Polar Vortex Prebreakup

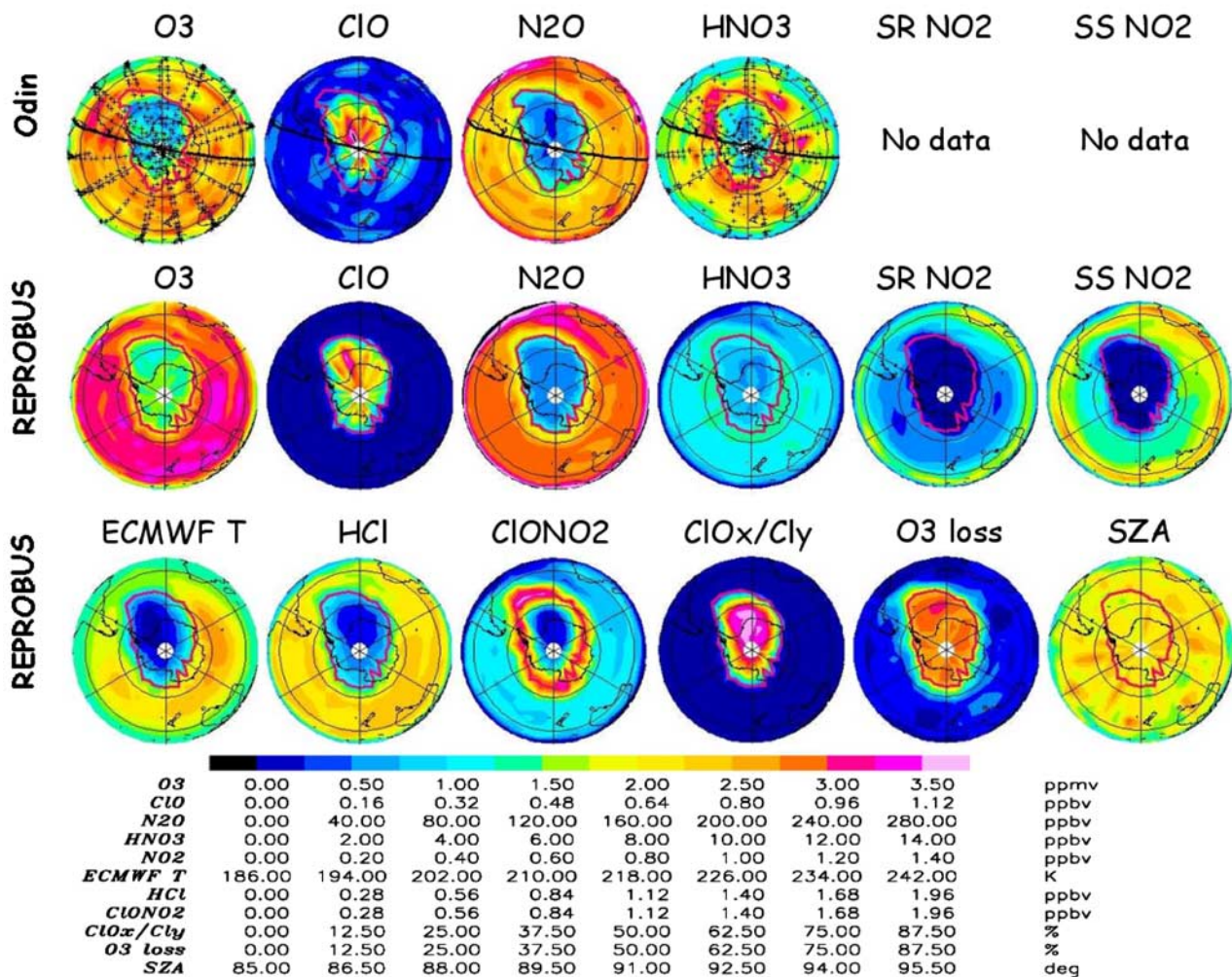
[10] N<sub>2</sub>O is a long-lived species in the lower stratosphere and is consequently a good tracer of dynamical motion. As a result of the subsidence, which started at the beginning of the winter, and the negative gradient of N<sub>2</sub>O with height, the Odin measurements at 500 K show very low mixing ratios in the polar vortex on 19–20 September (Figure 1). Indicated in red on each plot is the 120 ppbv N<sub>2</sub>O isopleth which coincides well at 500 K and during the period studied in this work to the maximum gradient of potential vorticity representative of the vortex edge. On 19–20 September, it can be seen from that criterion that the vortex is detected to be relatively symmetric and pole centered. This is consistent with meteorological analyses that indicate a compact, stable, and circular vortex [Simmons *et al.*, 2003], with temperatures well below 195 K on these two days. The REPROBUS N<sub>2</sub>O field is in good agreement with SMR in terms of horizontal extension of the polar vortex at 500 K but tends to overestimate slightly N<sub>2</sub>O at midlatitudes. This results from a too strong upward vertical transport above the Equator when using ECMWF analysis to drive the model, bringing too large N<sub>2</sub>O amounts to the tropical midstratosphere. This bias then propagates to midlatitudes by the descending branch of the Brewer-Dobson circulation and the quasi-horizontal transport of N<sub>2</sub>O from the tropics.

[11] The compactness of the vortex at this time of year is also visible from the observed vertical distribution of N<sub>2</sub>O (Figure 2). A strong horizontal gradient is visible across the vortex edge below 30 km altitude, with low N<sub>2</sub>O amounts over Antarctica, revealing a pronounced subsidence inside the vortex. The computed vertical distribution shows that this strong diabatic descent is well reproduced by REPROBUS in the core of a compact vortex well isolated from extra-vortex air.

[12] The O<sub>3</sub> field measured by Odin at 500 K shows a distinct minimum inside the vortex, with values lower than 1 ppmv. This is also shown by REPROBUS, but the model tends to overestimate the O<sub>3</sub> absolute amount compared to SMR. A 21% difference (1.5 versus  $1.2 \pm 0.1$  ppmv) is found in vortex average. The cumulative O<sub>3</sub> chemical loss since the beginning of the REPROBUS simulation can be obtained by calculating the difference between the chemically integrated ozone and a passive tracer initialized in the same way as ozone on 1 April 2002. On 19–20 September, the model indicates a O<sub>3</sub> chemical loss of the order of 70% at 500 K. Along the vertical (Figure 2), SMR O<sub>3</sub> retrievals



# 19–20 September 2002

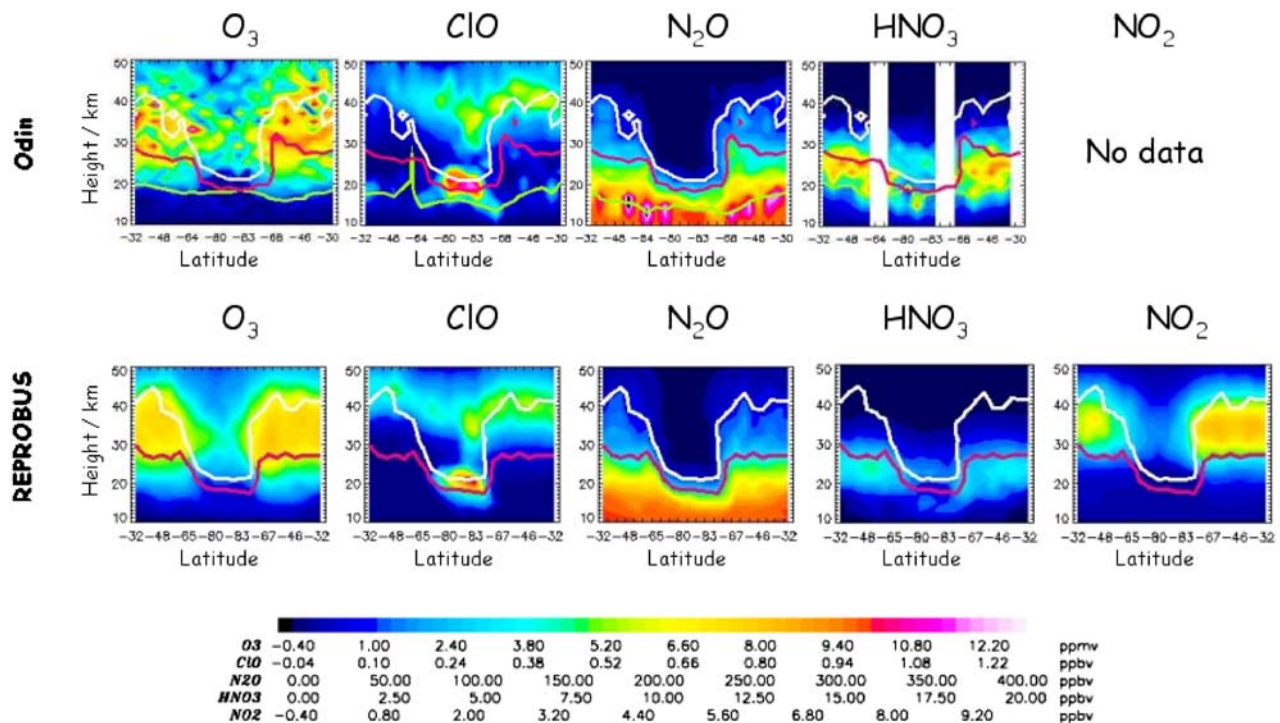


**Figure 1.** (top) From left to right,  $\text{O}_3$ ,  $\text{ClO}$ ,  $\text{N}_2\text{O}$ , and  $\text{HNO}_3$  fields measured by Odin/SMR on 19–20 September 2002, interpolated onto the 500-K potential temperature surface. The maps are polar orthographic projections with black circles at  $30^\circ\text{S}$  and  $60^\circ\text{S}$ . Crosses represent the Odin orbit tracks over the 24-hour period starting from 1200 UT. Locations of  $\text{ClO}$  and  $\text{N}_2\text{O}$  measurements are the same as  $\text{O}_3$  and are not represented. The red line corresponds to the 120-ppbv  $\text{N}_2\text{O}$  contour, which can be considered as the approximate edge of the polar vortex. The black line indicates the orbit track selected on Figure 2. Blank spaces represent data gaps or bad data points. Note that Odin measurements were performed at twilight ( $90^\circ < \text{SZA} < 92.5^\circ$ ) on 19–20 September 2002 and during daytime ( $\text{SZA} \sim 85^\circ$ ) for the other days. (middle) From left to right,  $\text{O}_3$ ,  $\text{ClO}$ ,  $\text{N}_2\text{O}$ ,  $\text{HNO}_3$ , and sunrise (SR) and sunset (SS)  $\text{NO}_2$  fields calculated by the REPROBUS chemical transport model on 19–20 September 2002, interpolated onto the 500-K potential temperature surface. Calculations are coincident in time and space with the Odin measurements. (bottom) From left to right, ECMWF temperature,  $\text{HCl}$ ,  $\text{ClONO}_2$ ,  $\text{ClO}_x/\text{Cl}$  ratio,  $\text{O}_3$  chemical loss calculated by REPROBUS (see text), and the solar zenith angle (SZA) at the time of the Odin measurements.

show that the depletion extends up to about 25 km. The large increase in  $\text{ClO}$  that is usually observed in the polar vortices in winter and spring is clearly detected by SMR. Even in the twilight conditions of the measurements ( $\text{SZA}$  range of  $90^\circ$ – $92.5^\circ$ ) on 19–20 September 2002, high values of  $\text{ClO}$  (0.5–1 ppbv) fill almost the entire vortex at 500 K. A good general agreement is observed between SMR and REPROBUS, with a vortex-averaged difference of 9% (0.70

versus 0.64 ppbv) at 500 K. This result reveals that chlorine activation processes are well reproduced by the model. A near complete removal of  $\text{HCl}$  and  $\text{ClONO}_2$  is computed in the vortex core. At the edge of the vortex, the  $\text{ClONO}_2$  field exhibits the typical collar structure that has often been observed at this time of year [e.g., Roche *et al.*, 1993]. From the assumption of photochemical equilibrium between  $\text{ClO}$  and its dimer  $\text{Cl}_2\text{O}_2$ , we calculate that the maximum

# 19–20 September 2002



**Figure 2.** (top) From left to right, latitude-height distribution of Odin/SMR  $O_3$ , ClO,  $N_2O$ , and  $HNO_3$  on 19–20 September 2002 along the orbit selected in Figure 1. Measurements are shown for latitudes southward of  $30^\circ S$  and were linearly interpolated onto a vertical grid of 2 km resolution. Each vertical tick on the x axis corresponds to one measured profile. The southernmost latitude is  $85^\circ S$  and is located at the center of the plot. The red line corresponds to the 120-ppbv  $N_2O$  contour also shown in Figure 1, and the white line corresponds to the 40-ppbv  $N_2O$  contour chosen to highlight the diabatic descent inside the vortex. The green line represents the lowermost altitude of significant measurements. Note that the plots gather sunrise measurements on the left-hand side and sunset measurements on the right-hand side. This is particularly important for  $NO_2$ , and to a lesser extent for midstratospheric ClO, which have an asymmetrical diurnal variation. (bottom) From left to right, coincident latitude-height distribution of  $O_3$ , ClO,  $N_2O$ ,  $HNO_3$ , and  $NO_2$  fields calculated by the REPROBUS chemical transport model.

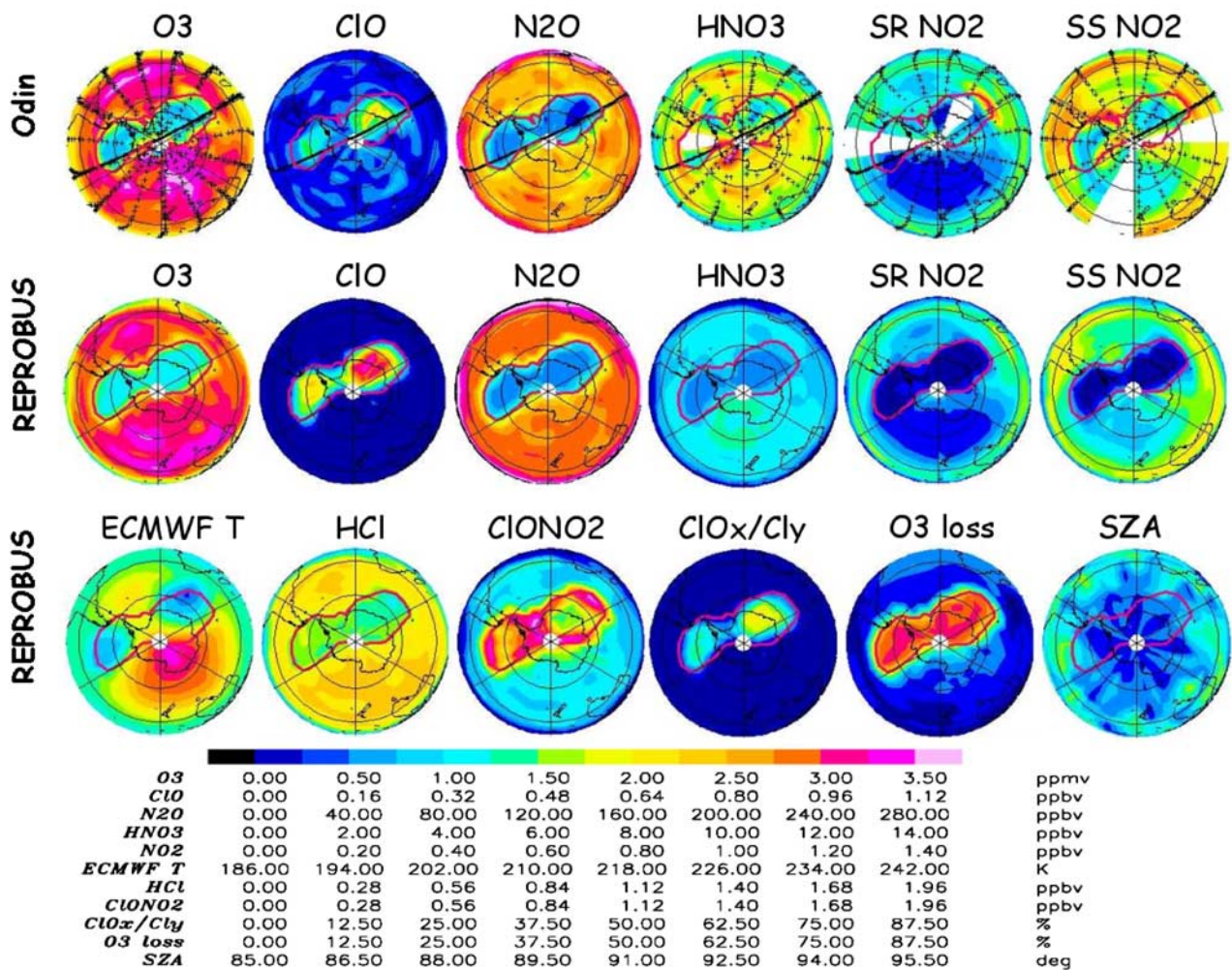
twilight ClO mixing ratios measured by SMR on 19–20 September correspond to a daytime ClO amount close to 2 ppbv. This is in good agreement with the largest values measured at 475 K inside the polar vortex by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) satellite instrument during the same period [Glatthor *et al.*, 2004]. We define the chlorine activation ratio as  $ClO_x/Cl_y$ , with  $ClO_x = Cl + ClO + 2Cl_2O_2$  and  $Cl_y$  as the total inorganic chlorine. On 19–20 September this ratio is calculated by REPROBUS to be about 85% at 500 K. Chlorine activation is observed between 19 and 25 km inside the vortex and correlates with the altitude range where  $O_3$  is depleted. In the midstratosphere, the ClO observed by Odin is maximum at about 40 km at midlatitudes and goes down to about 35 km inside the vortex as a result of subsidence. This is consistent with earlier observations from the ground in Antarctica [Emmons *et al.*, 1995; De Zafra *et al.*, 1995; Solomon *et al.*, 2002].

[13] The nominal character of the 2002 ozone hole until 19–20 September is reinforced by the SMR observations

of  $HNO_3$ , which show a distribution similar to that measured in previous Antarctic winters by the Microwave Limb Sounder (MLS) satellite-borne instrument [Santee *et al.*, 1999]. Low  $HNO_3$  mixing ratios (less than 5 ppbv) are observed by SMR in the polar vortex (Figure 1), while just outside of the vortex  $HNO_3$  values at 500 K are greater than 10 ppbv. Aerosol measurements by the Polar Ozone and Aerosol Measurement (POAM) satellite-borne instrument indicate that PSCs had nearly disappeared at the latitude of the observations (poleward of  $85^\circ S$ ) on 20 September [Nedohula *et al.*, 2003]. This is also supported by REPROBUS, which calculates that the maximum amount of condensed  $HNO_3$  at 500 K (not shown) is only of the order of 0.5 ppbv, and is confined to a limited area above the Weddell Sea where the ECMWF temperature is lowest. Thus it is likely that the low  $HNO_3$  mixing ratios observed by Odin on 19–20 September inside the vortex are due to irreversible loss by sedimentation of  $HNO_3$ -containing PSC particles, rather than to temporary removal from the gas phase by



## 25-26 September 2002



**Figure 3.** Same as Figure 1 but for the 25–26 September 2002 period. In Figure 3 (top), O<sub>3</sub>, sunrise (SR) and sunset (SS) NO<sub>2</sub> fields are measured by OSIRIS.

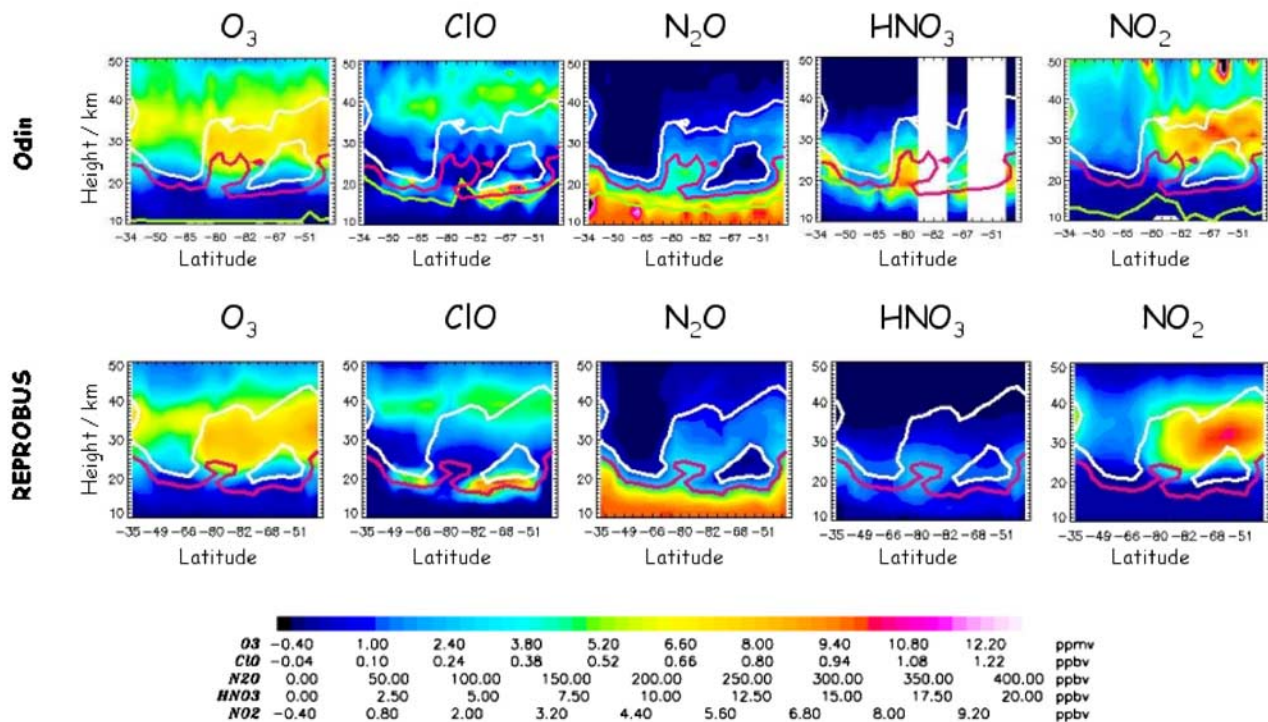
sequestration in cloud particles. Outside the polar vortex, the model underestimates the HNO<sub>3</sub> amount by almost a factor of two, a result also supported by the comparison of REPROBUS to preliminary HNO<sub>3</sub> observations by the MIPAS instrument (not shown). This bias is again related to the too fast vertical transport of N<sub>2</sub>O in the tropical midstratosphere, resulting in too weak production of NO<sub>y</sub> in the model.

#### 4.2. Polar Vortex Breakup

[14] During the last week of September 2002, meteorological analyses reveal the development of a Southern Hemisphere major stratospheric warming that results in a strong dynamical disturbance of the polar vortex [Manney *et al.*, 2005]. On 25–26 September the elongated shape of the polar vortex at 500 K is evident in the N<sub>2</sub>O distribution measured by the SMR (Figure 3). REPROBUS also reproduces the horizontal extension of the vortex toward midlatitudes. From a consideration of the measurements in the orbit plane (Figure 4), it is evident that extra-vortex air,

characterized by larger values of N<sub>2</sub>O, is present above the pole between 20 and 25 km altitude. Further, the SMR observations show that the vortex structure is not as vertically uniform as on 19–20 September 2002: there is an N<sub>2</sub>O-rich layer above the low N<sub>2</sub>O values measured in the vortex lobe extending toward the Indian Ocean that clearly indicates the presence of midlatitude air above 30 km. The model outputs reveal intrusion of extra-vortex air above polar latitudes in good agreement with SMR. Thus dynamics seems to be well reproduced by the model during such perturbed conditions, even though the model still overestimates N<sub>2</sub>O at high altitudes. The split of the vortex is also visible from the OSIRIS ozone measurements that also show ozone-rich air being transported toward the pole above the vortex, in agreement with Hoppel *et al.* [2003]. At 500 K the ozone distribution mimics the shape of the N<sub>2</sub>O field with two ozone-poor elongated lobes. It should be noted that the O<sub>3</sub> mixing ratios in the polar vortex on 25–26 September do not show a significant reduction compared to those measured

## 25–26 September 2002



**Figure 4.** Same as Figure 2 but for the 25–26 September 2002 period. In Figure 4 (top),  $O_3$  and  $NO_2$  fields are measured by OSIRIS.

5 days earlier. This suggests a reduced  $O_3$  chemical loss between these two dates. Continuing  $O_3$  destruction might be compensated by increased mixing with  $O_3$ -rich midlatitude air during the warming event. However, the mixing would be accompanied by an increase in  $N_2O$  values between 19–20 and 25–26 September, which is not unambiguously observed by Odin.

[15] On 25–26 September the ClO measurements by the SMR were made in daytime at all latitudes (SZA range of  $85^{\circ}$ – $88^{\circ}$ ). The maximum ClO mixing ratios are about 0.7 ppbv at 500 K. This corresponds to a  $\sim 70\%$  reduction relative to the estimated daytime ClO on 19–20 September, in agreement with the decrease in ClO measured by MIPAS after 20 September [Glatthor *et al.*, 2004]. The vortex averaged ClO mixing ratio measured by SMR on 25–26 September ( $0.35 \pm 0.03$  ppbv) is also in good agreement with the daily averages derived from MIPAS at 520 K on the same days (0.25 and 0.40 ppbv on 25 and 26 September, respectively). The decline in ClO is a typical feature of the mid to late September period in Antarctica, as shown by the 6-year climatology derived from the MLS satellite measurements [Santee *et al.*, 2003], although MLS could never observe the complete chlorine recovery period. Ground-based observations performed in Antarctica in 1992 [Emmons *et al.*, 1995], in 1993 [De Zafra *et al.*, 1995], and over the 1996–2000 period [Solomon *et al.*, 2002] also gave evidence of the chlorine deactivation that occurs during the third week of September. Daytime ClO mixing ratios at 20 km were found to be in general smaller than

1 ppbv around 25–26 September, which is consistent with the SMR observations in 2002.

[16] It must be noted that the model ClO values on 25–26 September are larger than those measured by SMR: the vortex averaged ClO is overestimated by 60%. Thus the actual rate of chlorine deactivation was faster than calculated by the model. This can be related to the underestimation of  $NO_2$  in the vortex by REPROBUS (see Figure 3), which does not allow a conversion of ClO to  $ClONO_2$  as fast as detected by SMR. This result is very sensitive to the absolute amount of  $NO_y$  available in the polar vortex. In a simulation for which the initial  $HNO_3$  mixing ratio at 500 K is increased by 1.5 ppbv relative to the reference experiment presented here, the substantially faster deactivation rate that is calculated by the model reduces by a factor of three the overestimation of ClO on 25–26 September. In addition to this effect, the slight model overestimation of  $O_3$  may not have favored the HCl recovery, which is known to be inversely proportional to the absolute amount of  $O_3$  [see, e.g., Douglass *et al.*, 1995].

[17] A rapid rise of the lower stratospheric temperatures occurred during the 2002 vortex split episode: On 25–26 September, minimum ECMWF temperatures are above 195 K at most altitudes (Figure 3), i.e., above the threshold for PSC formation. The absence of PSCs is supported by the POAM [Nedohula *et al.*, 2003] and OSIRIS (D. Gattinger, personal communication) aerosol measurements, which do not exhibit high extinction coefficients for these two days. This is in contrast to years prior to 2002, when PSCs were



## 1-2 October 2002

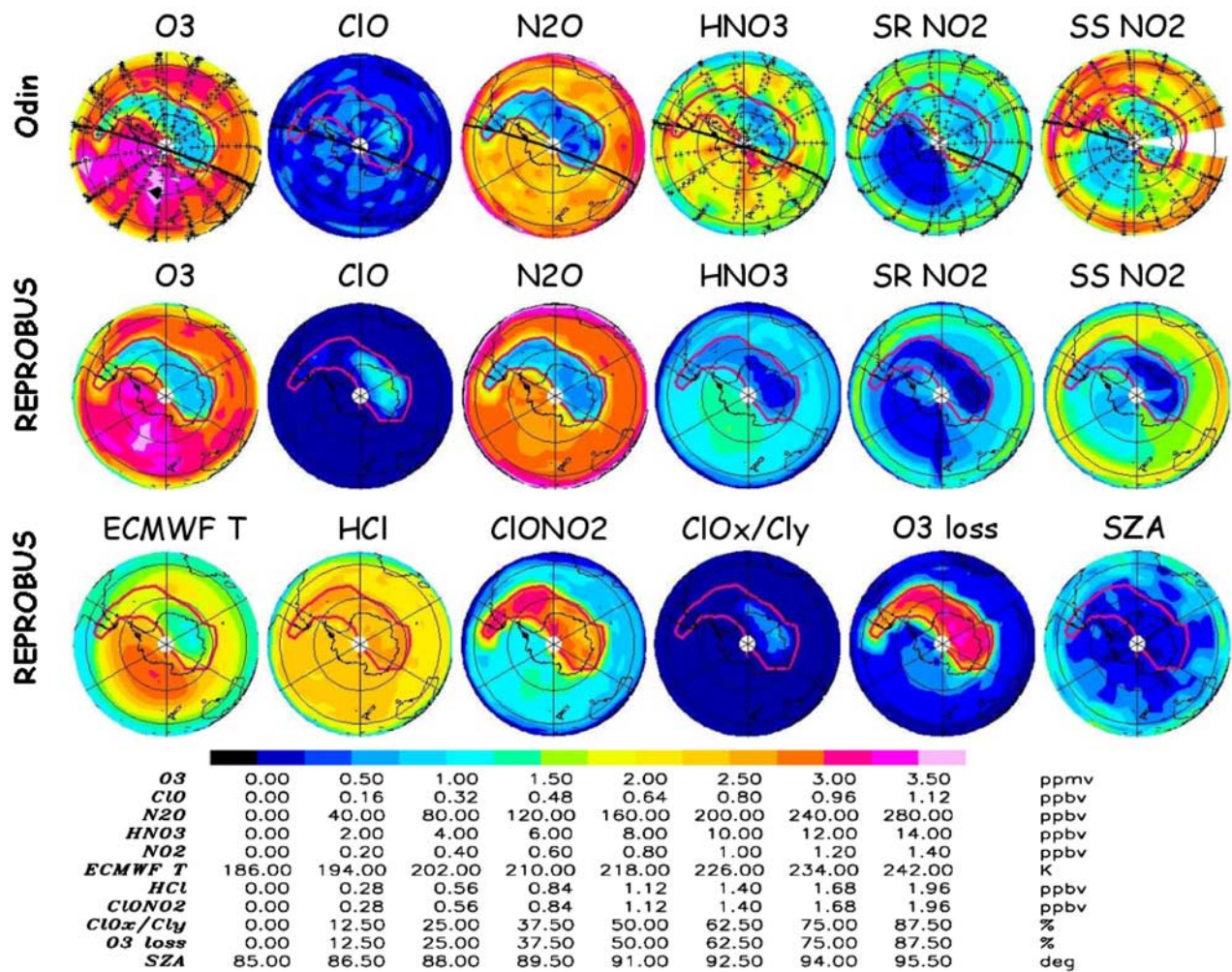


Figure 5. Same as Figure 3 but for the 1–2 October 2002 period.

usually observed until the middle of October [Poole and Pitts, 1994; Fromm *et al.*, 1997]. Despite the absence of PSCs, it is important to note that the  $\text{HNO}_3$  distribution measured by SMR on 25–26 September still shows a marked contrast between the vortex ( $\text{VMR} < 5$  ppbv) and midlatitude air ( $\text{VMR} > 8$  ppbv) although few  $\text{HNO}_3$  measurements are available at 500 K for that period. This confirms that the low  $\text{HNO}_3$  mixing ratios detected in the vortex are the result of irreversible loss (or denitrification) of nitric acid via the earlier sedimentation of PSC particles. The in-vortex  $\text{HNO}_3$  values measured by SMR at 500 K (2–4 ppbv) are in good agreement with ground-based measurements [e.g., De Zafra and Smyshlyaev, 2001] for this time of year. They are also consistent with the  $\text{HNO}_3$  climatology established from 6 years of data by the MLS satellite instrument [Santee *et al.*, 1999].

[18] On 25–26 September,  $\text{NO}_2$  was measured by OSIRIS at sunrise (SR) and sunset (SS). Outside the vortex, OSIRIS observations show the expected increase in  $\text{NO}_2$  during the day. Over the Pacific Ocean, the large-scale  $\text{NO}_2$  minimum correlates well with elevated  $\text{O}_3$  and high temperatures. This is consistent with the standard gas-phase

photochemical theory represented at midlatitudes by REPROBUS. Inside the polar vortex, OSIRIS shows low values of  $\text{NO}_2$  and a limited increase during the day, in agreement with the model. The removal of the nitrogen oxides in the polar vortex results from their conversion to reservoir forms of nitrogen during the polar night and from denitrification. However, the calculated  $\text{NO}_2$  underestimate the measured values in the two vortex lobes, a rather typical bias of photochemical models in wintertime polar latitudes [Payan *et al.*, 1999]. In the orbit plane (Figure 4), the diurnal variation of  $\text{NO}_2$  is clearly visible above 25 km in the OSIRIS measurements. The main features of the  $\text{NO}_2$  vertical distribution and the diurnal variation measured by OSIRIS are well reproduced by REPROBUS.

#### 4.3. Polar Vortex Postbreakup

[19] On 1–2 October 2002 the  $\text{N}_2\text{O}$ -poor air representative of the vortex reveals that the vortex is displaced from the pole with a large elongation over South America well reproduced by the model (Figure 5). In the orbit plane chosen in Figure 6, the main vortex is clearly detected from the  $\text{N}_2\text{O}$  field around 87S with its elongation, centered on

## 1-2 October 2002

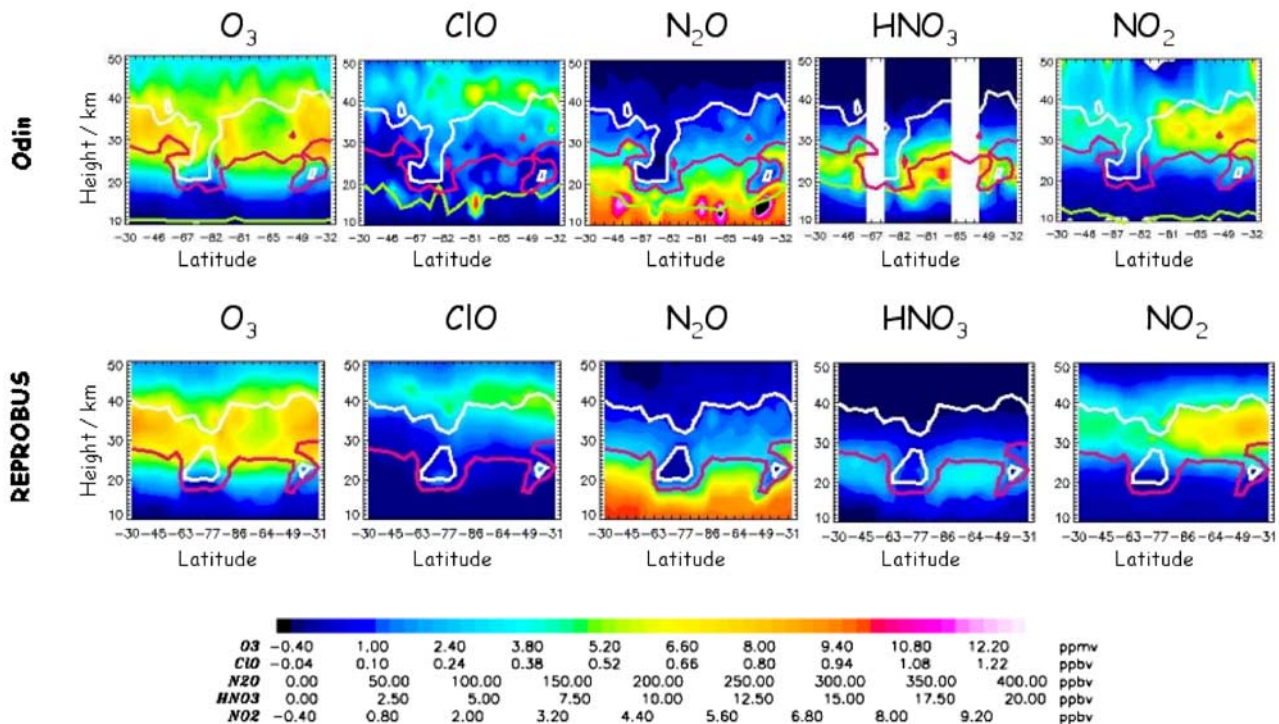


Figure 6. Same as Figure 4 but for the 1–2 October 2002 period.

49S, only visible in the lowermost stratosphere. The O<sub>3</sub>-depleted area correlates very well with the region of low N<sub>2</sub>O. The vortex averaged ozone mixing ratio ( $0.99 \pm 0.02$  ppmv) is nearly identical to that measured on 25–26 September ( $1.01 \pm 0.02$  ppmv), this indicates that little further loss occurred at 500 K after that date. In the model, the too large ClO amounts that were still present on 25–26 September have yielded to an additional O<sub>3</sub> loss peaking at 80% on 1–2 October. The signature of the ozone hole is also detected far from the Antarctic region, with O<sub>3</sub> mixing ratios lower than 1.5 ppmv above southern Chile and Argentina. A striking feature of the Odin daytime observations on 1–2 October is that ClO is close to the SMR detection limit (0.2 ppbv) inside the vortex. REPROBUS also calculates a rapid ClO decrease, although the maximum ClO amounts (0.3 ppbv) are still slightly larger than measured by SMR. The chlorine activation ratio passed from  $\sim 50\%$  to less than  $\sim 15\%$  in 5 days. In the orbit plane, Figure 6 shows that the vortex is free of enhanced ClO at all vertical levels below 30 km. Atmospheric chlorine has therefore evolved from a situation of nearly complete activation to a situation of complete recovery in less than 10 days. This change appears to be faster than the only continuous observations of ClO available over the late September to early October period, performed from the Scott Base (Antarctica, 78°S) by Solomon *et al.* [2002] in 1996 and 2000.

[20] On 1–2 October, in the elongated part of the vortex at latitudes northward of 60°S, OSIRIS measurements show a substantial increase of SS NO<sub>2</sub>, with values larger than 1 ppbv. SMR gives an indication of elevated abundances of

HNO<sub>3</sub> in this region, which must be considered with caution as only three profiles are available in the vortex tongue that extends over the tip of South America. Nevertheless, it is likely that the displacement from the pole and the elongated shape toward midlatitudes favored the exposure to unusually strong solar irradiance at this time of the year so leading to the early photolysis (and oxidation by OH) of HNO<sub>3</sub> and the subsequent enhanced production of NO<sub>2</sub>. Even with HNO<sub>3</sub> amounts lower than those measured by SMR, the model indicates that the HNO<sub>3</sub> photolysis and oxidation are fast enough to titrate entirely ClO in the vortex tongue, so leading to a substantial ClONO<sub>2</sub> buildup in the same area (Figure 5). Thus Odin and REPROBUS both indicate that the unusual morphology of the Antarctic vortex may have been an important cause for the acceleration of the chlorine deactivation in late September to early October 2002.

[21] On 4–5 October 2002 the N<sub>2</sub>O field at 500 K (Figure 7) indicates that the core of the polar vortex is returning to a more typical behavior, centered over the Antarctic continent. A filament extends over South America where REPROBUS calculates a cumulative ozone loss of about 40%. From the measured and simulated N<sub>2</sub>O vertical structure (Figure 8), it is evident that the polar vortex is far from vertically uniform. About half of the vortex is observed in the form of a  $\sim 3$ -km-thick slice in the lower stratosphere, overlaid by midlatitude air extending up to the stratopause. The “boot shape” of the polar air visible in Figure 8 is also clearly visible in the vertical distributions of ozone and nitric acid.

[22] It is also noteworthy that the O<sub>3</sub> and N<sub>2</sub>O values measured at 500 K have not increased in the main polar



## 4-5 October 2002

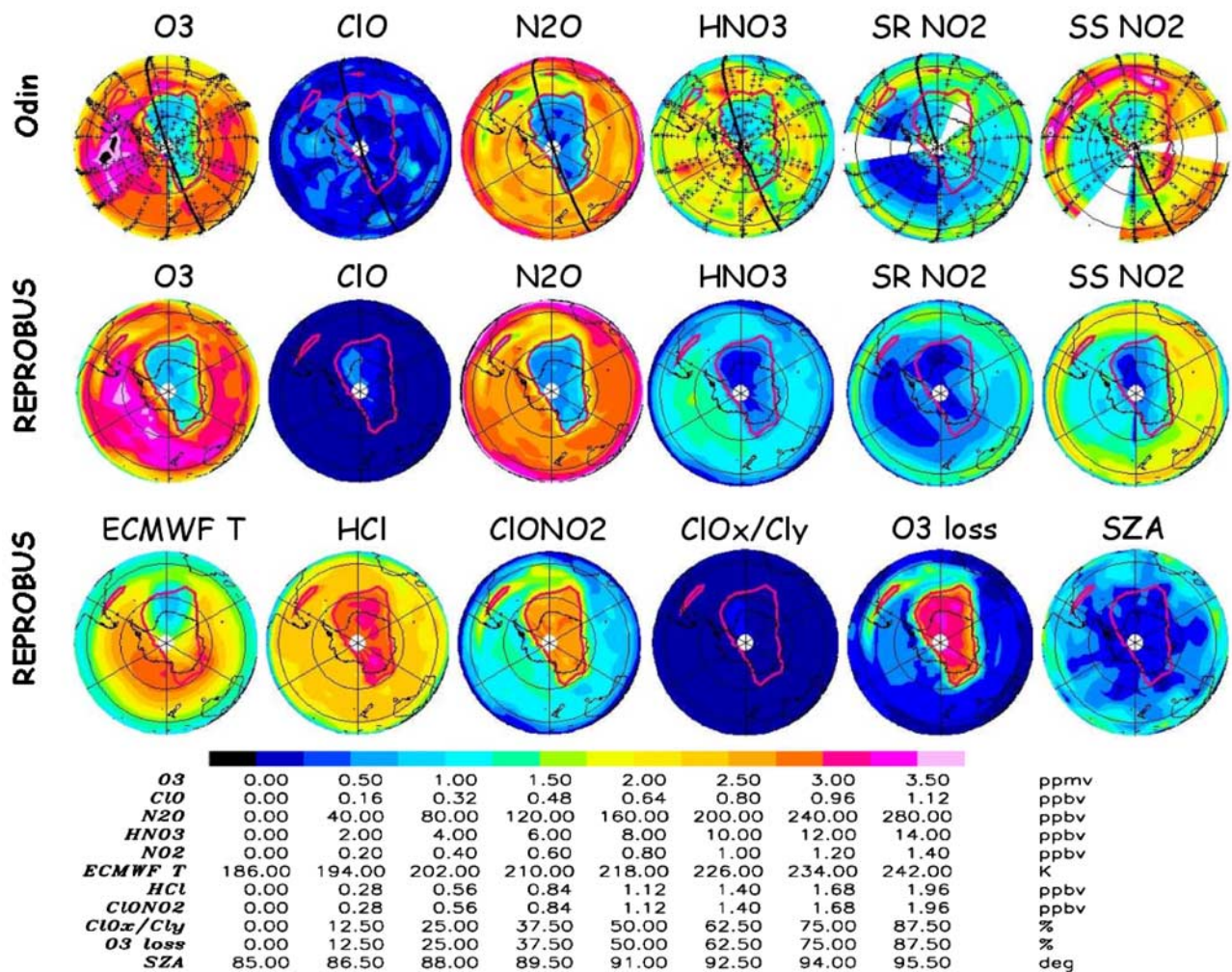


Figure 7. Same as Figure 3 but for the 4–5 October 2002 period.

vortex since 19–20 September. This supports the idea of a relatively isolated vortex during the whole period, and that mixing with midlatitude air did not play a major role in the observed deactivation of chlorine. In contrast, in situ production of  $\text{NO}_x$  is suggested by the measured  $\text{NO}_2$  distribution showing a gradual increase in the region of the vortex where  $\text{HNO}_3$  is less affected by denitrification. Daytime ClO is maintained at a level below the SMR detection limit. REPROBUS results show very low values of ClO, in good agreement with the observations, and the chlorine activation ratio calculated by the model is close to zero.

## 5. Conclusions

[23] The SMR and OSIRIS instruments onboard the Odin satellite have measured stratospheric constituents such as  $\text{O}_3$ , ClO,  $\text{N}_2\text{O}$ ,  $\text{HNO}_3$  and  $\text{NO}_2$  during the 2002 Antarctic major warming. These observations enabled the study of the dynamical and chemical evolutions of the polar vortex at and above the 500 K isentropic surface. On 19–20 September the Antarctic vortex was dynamically stable and

chemically nominal with a nearly complete activation of chlorine and ozone amounts consistent with a 70% chemical loss on the 500 K isentropic surface. Our 3-D model simulation suggests that the low  $\text{HNO}_3$  mixing ratios measured by Odin inside the polar vortex result from earlier denitrification. On 25–26 September, Odin  $\text{N}_2\text{O}$  observations reveal the unusual morphology of the Southern Hemisphere vortex associated to a stratospheric warming, signature of the vortex split episode. Observations show a decrease in ClO that is quantitatively consistent with measurements performed during the same period in 2002 and in the previous years. On 1–2 October a nearly complete deactivation of the ClO radicals is observed, translating into the end for the ozone-destroying chemical cycles. This acceleration of the chlorine deactivation results both from the warming of the Antarctic vortex in 2002, putting an early end to the polar stratospheric cloud season, and from the vortex elongation toward regions of strong solar irradiance, which is shown by our model simulation to have favored the rapid reformation of  $\text{ClONO}_2$ . Once the vortex split episode is over, the  $\text{O}_3$  field is measured to be stable within the vortex while  $\text{NO}_2$  gradually increases. The



## 4-5 October 2002

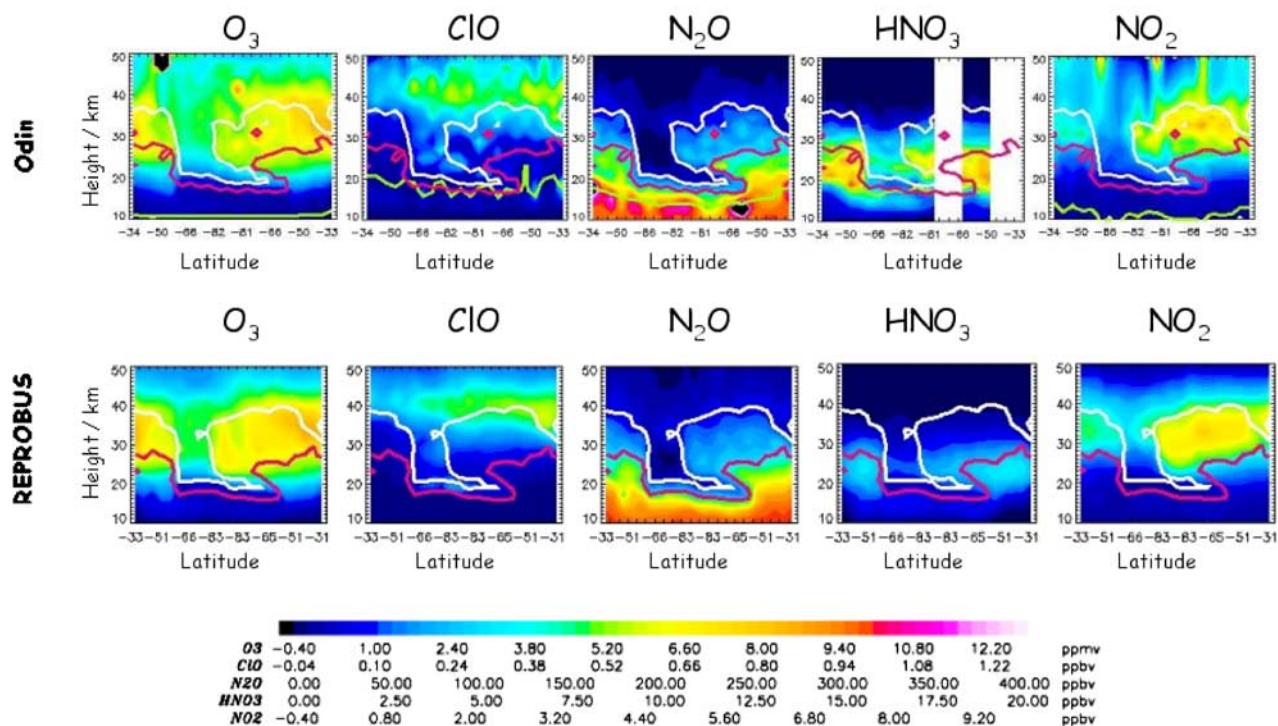


Figure 8. Same as Figure 4 but for the 4–5 October 2002 period.

dynamical and chemical evolution of the vortex is qualitatively well reproduced by the model. Quantitative differences are shown to be mainly attributable to the too weak amounts of  $HNO_3$  in the model, which do not produce enough  $NO_2$  in presence of sunlight to deactivate  $ClO$  as fast as observed by Odin in 2002.

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